

# Low-Temperature Growth of DKDP for Improving Laser-Induced Damage Resistance at 350 nm

*A. K. Burnham, M. Runkel, R. A. Hawley-Fedder, M. L.  
Carman, R. A. Torres, P. K. Whitman*

This article was submitted to  
32<sup>nd</sup> Annual Symposium on Optical Materials for High Power Lasers  
Boulder, CO  
October 16-18, 2000

**U.S. Department of Energy**

Lawrence  
Livermore  
National  
Laboratory

**December 6, 2000**

## DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

This report has been reproduced directly from the best available copy.

Available electronically at <http://www.doc.gov/bridge>

Available for a processing fee to U.S. Department of Energy  
And its contractors in paper from  
U.S. Department of Energy  
Office of Scientific and Technical Information  
P.O. Box 62  
Oak Ridge, TN 37831-0062  
Telephone: (865) 576-8401  
Facsimile: (865) 576-5728  
E-mail: [reports@adonis.osti.gov](mailto:reports@adonis.osti.gov)

Available for the sale to the public from  
U.S. Department of Commerce  
National Technical Information Service  
5285 Port Royal Road  
Springfield, VA 22161  
Telephone: (800) 553-6847  
Facsimile: (703) 605-6900  
E-mail: [orders@ntis.fedworld.gov](mailto:orders@ntis.fedworld.gov)  
Online ordering: <http://www.ntis.gov/ordering.htm>

OR

Lawrence Livermore National Laboratory  
Technical Information Department's Digital Library  
<http://www.llnl.gov/tid/Library.html>

# Low-temperature growth of DKDP for improving laser-induced damage resistance at 350 nm

A. K. Burnham,\* M. Runkel, R. A. Hawley-Fedder, M. L. Carman, R. A. Torres,  
and P. K. Whitman

Lawrence Livermore National Laboratory  
P.O. Box 808, L-487 Livermore, CA 94551  
Phone: (925)-422-7304, burnham1@llnl.gov

## ABSTRACT

A set of twenty-three 20-L crystallizer runs exploring the importance of several engineering variables found that growth temperature is the most important variable controlling damage resistance of DKDP over the conditions investigated. Boules grown between 45°C and room temperature have a 50% probability of 3 $\omega$  bulk damage that is 1.5 to 2 times higher than boules grown between 65 and 45°C. This raises their damage resistance above the NIF tripler specification for 8 J/cm<sup>2</sup> operation by a comfortable margin. Solution impurity levels do not correlate with damage resistance for iron less than 200 ppb and aluminum less than 2000 ppb. The possibility that low growth temperatures could increase damage resistance in NIF-scale boules was tested by growing a large boule in a 1000-L crystallizer with a supplemental growth solution tank. Four samples representing early and late pyramid and prism growth are very close to the specification as best it is understood at the present. Implications of low temperature growth for meeting absorbance, homogeneity, and other material specifications are discussed.

**Keywords:** KDP, DKDP, frequency conversion crystals, laser damage

## 1. INTRODUCTION

Although the NIF baseline construction plan is to use conventional growth DKDP for triplers for at least two clusters, the long-term flexibility and cost advantages of rapid growth material make it an object of continued research. One of the issues with rapid growth DKDP is its resistance to laser-induced bulk damage at 351-nm (3 $\omega$ ).<sup>1</sup> The 351-nm bulk damage resistance of previous, rapidly grown DKDP has been marginal for long-term operation of the NIF at an average fluence of 8 J/cm<sup>2</sup> over 3 ns. For example, the 10% damage probability of tripler-cut samples from Beamlet boule RGA8 ranged from 9.5 to 11.5 J/cm<sup>2</sup> at 7.6 ns, compared to a proposed specification of 12.5 J/cm<sup>2</sup> at 7.6 ns. As a result, there has been a significant effort over the past two years to improve the quality of rapid growth DKDP to meet NIF performance goal of < 0.1% scattering loss from bulk damage at 8 J/cm<sup>2</sup> average fluence. This paper presents data suggesting that rapid growth starting at saturation temperatures of about 45°C should meet that specification, although there may be scaling issues for solution volume to filtration rate, based on initial results from NIF-scale boules grown in 1000-L crystallizers, and newly emerging concerns about surface crazing.

## 2. APPROACH

Small (~6") crystal boules characterizing a range of conditions were grown on a point seed using the temperature reduction technique in two standard 20-liter Holden-type crystallizers.<sup>2</sup> Growth parameters were chosen to explore the obvious engineering variables that can be controlled during rapid growth production. The parameters were initial salt purity (within the range of current production salts), resaturation sequence, crystallizer tank material (glass and polycarbonate), filter pore size in the constant filtration unit, and growth temperature (65 to 45°C and 45 to 25°C). In all cases, previous work indicated that these parameters are important at some level: addition of iron phosphate decreases damage resistance (M. Yan, M. Runkel, M. Staggs, and J. De Yoreo, unpublished results, 1999), constant filtration improves damage resistance,<sup>3,4</sup> glass

dissolves at a finite rate in the growth solution,<sup>5</sup> and inspection of earlier limited, unpublished data sets suggested temperature might be important.

Five-cm square by 1-cm thick samples were cut from the boules in two orientations: (1) optical axis parallel to the pyramidal direction (z axis), otherwise known as z-cuts, and (2) type-II frequency conversion orientation with the optical axis tipped at 59° relative to the pyramidal direction, otherwise known as tripler cuts. Z-cuts were made from all crystals. During the course of the work, it was discovered that the damage resistance of z cuts is much higher than tripler cuts, which are the orientation of interest.<sup>6</sup> Consequently, a subset of triplers was then fabricated for testing. In both cases, the location of the damage samples was such that they are mostly pyramidal material.\* It is likely that the laser test grid often went through both prism and pyramid material. We have not found difference in damage resistance between pyramid and prism material in earlier testing.<sup>4,7</sup>

Damage resistance was measured on the Zeus damage tester, which uses a commercial, table-top Nd-system operating at 355 nm.<sup>8</sup> Damage is detected by a photomultiplier tube, which measures changes in signal scatter from a HeNe laser that is colinear with the 3ω-damage beam. Damage tests were of two types:

S/1—each of about 10 spots receives about 100 shots at a specified fluence

R/1—each of about 100 spots receives up to 100 shots ramped from low to high fluence until damage occurs

In the both cases, the probability of damage is plotted as a function of the test fluence. The R/1 probability curve is typically 1.5-2.0 times higher because of laser conditioning. In other words, as the fluence is ramped during the test, the defects initiating damage tend to dissipate, allowing the crystal to receive a higher fluence before a given level of damage occurs.

### 3. RESULTS

A summary table of the growth conditions and damage results is given in Table 1. The S/1 and R/1 curves are shown in Figures 1-4 for the four boules that are available both as z-cuts and triplers. The solid line in the figures is for sample DKDP-11, which represents our current estimate of acceptability for 8 J/cm<sup>2</sup> operation with <0.1% scattering from bulk damage.

Several conclusions are evident from Table 1 and Figures 1-4. First, the z-cut crystals have a systematically higher damage resistance than the tripler crystals, as has been seen for other crystal sets.<sup>6</sup> Second, the R/1 profiles are systematically higher than the S/1 profiles, again as has been seen before,<sup>9</sup> although the ratio is proportionately less for the more damage resistant crystals grown at the lower temperature interval. In other words, as the crystal quality improves the effect of laser conditioning becomes slightly less. Finally, and the main point of this paper, the crystals grown between 45 and 25°C are systematically more damage resistant than the crystals grown between 65 and 45°C. Excluding the crystals grown with added iron, the 50% S/1 damage probability ranges from 14-22 J/cm<sup>2</sup> for the high temperature crystals and 25-40 J/cm<sup>2</sup> for the low temperature crystals. Figure 5 shows the two clusters of S/1 damage distribution profiles for z-cuts of all the growth runs except the ones with iron doping. Figure 6 shows the relationship between the 50% damage probability and saturation temperature more clearly, and the iron-doped growth runs fall on the same trendline.

Various run comparisons can be used to show that the other variables besides growth temperature are not particularly important. The seemingly unimportant variables are iron and aluminum concentration, resaturation sequence, pore filter size, tank composition, and residual EDTA concentration.

First, because iron was located twice in analyses of damage craters and because the addition of multi-ppm levels of iron phosphate (relative to DKDP) causes a decrease in the damage fluence in earlier unpublished studies (J. De Yoreo et al., 1998), it has been proposed that insoluble iron phosphate, or some other type of particulate, serves as the initiation site for damage. Earlier work reasoned that homogeneous absorption does not create enough localized heating to cause damage and that damage must be due to a concentrated absorber such as an iron phosphate particle with a diameter of tens to hundreds of nm.<sup>4</sup> While it is clear that earlier work shows that iron phosphate can be a problem at concentrations greater than 1 ppm, it is

---

\* Sample #'s 688 and 701 were pyramidal material only. Sample #681 had two test plates, one prism and one pyramidal, that gave identical results. Sample #'s 715 and 746 were mostly prism material.

Table 1. DKDP Sample Growth Runs for 3 $\omega$  Damage Studies

Tank Mat'l	CFS Filter Size, $\mu\text{m}$	Starting Temp, $^{\circ}\text{C}$	Salt	Xtal No.	EDTA (mole %)	Solution Al, ppb (pre/post)	Solution Fe, ppb (pre/post)	z-cut 50% S/1	z-cut 50% R/1	tripler 50% S/1	tripler 50% R/1
Glass	0.02	60.5	05188	625*					27	12.5	18
Glass	0.02	42.8	05188	636		870/720	150/96	40	44	21	29.5
Glass	0.02	63.1	05188/06208	651		550/860	98/94	18	32		
Glass	0.006	46.3	06208/05188	694		1300/870	100/52	33			
Glass	0.006	42.9	06208	709		590/690	140/100	31	37	21	30
Polycarbonate	0.006	58.7	06208	715		ND/200	120/120	20	27.5	9.5	14.5
Polycarbonate	0.006	46.3	06208	727		290/D	130/61	33		25	
Polycarbonate	0.006	62.4	06208/03099	731	0.0168	D/350	63/88	18			
Polycarbonate	0.006	58.7	06208/03099	737	0.0244	390/480	91/120	21			
Polycarbonate	0.006	57.9	03099	746	0.0712	ND/310	61/76	19			
Glass	0.006	68.4	5156	757	0.0056	D/440	110/130	16			
Glass	0.006	57.8	5156	768	0.0057	590/640	2000/2100	18			
Glass	0.006	59.8	pyramid	668		ND/430	ND/D	18	24		
Glass	0.006	60.6	pyramid	681		590/700	D/D	18	30		
Glass	0.006	40.9	pyramid	701		2300/2000	190/140	25	30		
Glass	0.006	64.7	06208	710		D/570	97/96	17	28	7.5	15
Glass	0.006	43.8	06208	720		720/550	96/68	27		14	20
Glass	0.006	64.6	06208/03099	728	0.0116	460/750	78/100	20		7.5	15
Glass	0.006	57.1	06208	735	0.0130	950/680	110/150	22			
Glass	0.006	62.4	03099	745	0.0634	?/770	?/97	22			
Glass	0.006	63.2	5155	756	0.0018	270/500	79/96	14			
Glass	0.006	49.9	5155	767	0.0019	490/470	1100/990	22			
Glass	0.006	64.1	156272	778		D/460	200/200	18			

not clearly an issue at the levels present in the current production salts. Crystal #778 had the highest solution iron concentration (200 ppb) of any of the runs, yet its damage resistance is within the range of the other boules grown from 65 to 45 $^{\circ}\text{C}$ . The crystals with the next highest concentration, #636 (150 ppb) and #701 (190 ppb), have high damage profiles within the range of other crystals grown below 45 $^{\circ}\text{C}$ .

Resaturation sequence could conceivably cause a difference in damage resistance if the first growth run in a sequence cleans up either dissolved or particulate impurities related to damage by either filtration or incorporation into the first boule. However, low temperature boules from a fresh solution (#694 and #709) fall in the same damage resistance range as those from a first resaturation (#636, #727, #701, and #720).

A smaller pore size in the continuous filtration system could conceivably be more effective at eliminating particulates or clusters in the growth solution, and consequently in the boule. After three growth runs in one crystallizer, the polysulfone filters in both systems were changed from 0.02 to 0.006  $\mu\text{m}$  pore diameter. However, the low temperature boule using the 0.02  $\mu\text{m}$  filter (#636) has a slightly greater damage resistance than the other low temperature boules, and the high-temperature boule using the 0.02  $\mu\text{m}$  filter (#651) is the same as many other high-temperature boules grown with 0.006  $\mu\text{m}$  filters.

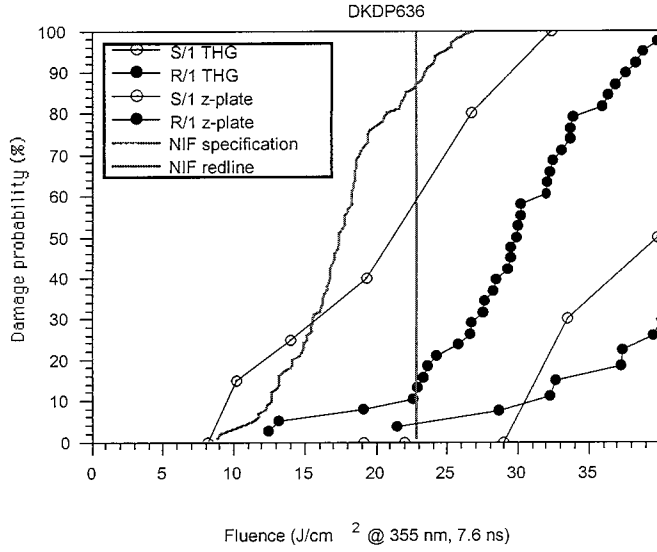


Figure 1. Unconditioned (S/1) and conditioned (R/1) damage curves for tripler and z-cut crystals from Low-T boule DKDP636.

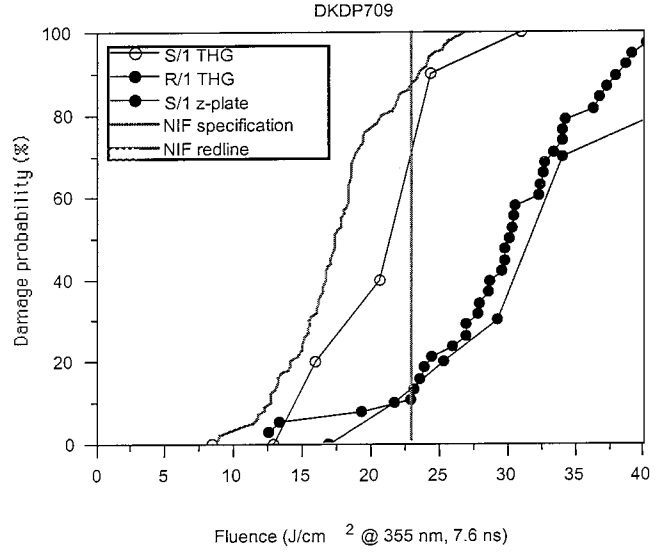


Figure 2. Damage curves from tripler and z-plates from low-T boule DKDP709.

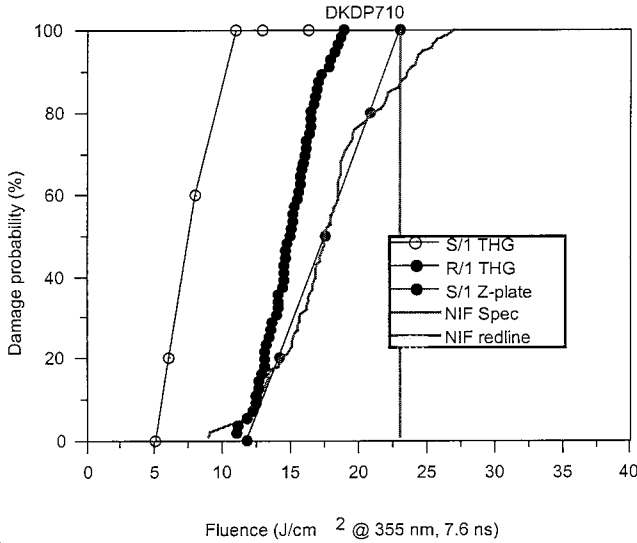


Figure 3. Damage curves from high-T boule DKDP710.

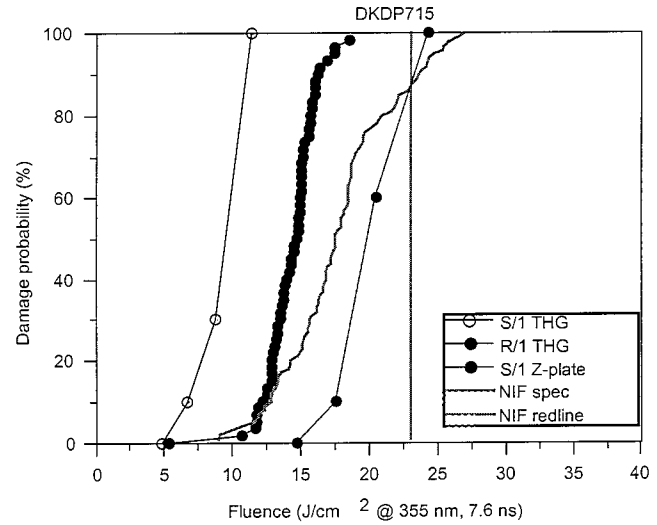


Figure 4. Damage curves from high-T boule DKDP715.

Tank material could potentially make a difference because of impurities that leach from the tanks. Pyrex slowly dissolves at a measurable rate in growth solutions, and trace organics could conceivably leach from polycarbonate. In particular, polycarbonate usually contains UV stabilizers, although the pedigree of our tank is not known. However, the one low temperature boule grown in polycarbonate had a high damage resistance, and high temperature boules grown in polycarbonate tanks had similar damage resistance to those grown in glass, so tank material does not seem to be important. A caveat in this comparison is that 20-L growth runs are short compared to 1000-L growth runs, so they have less time to accumulate impurities from tank dissolution or leaching.

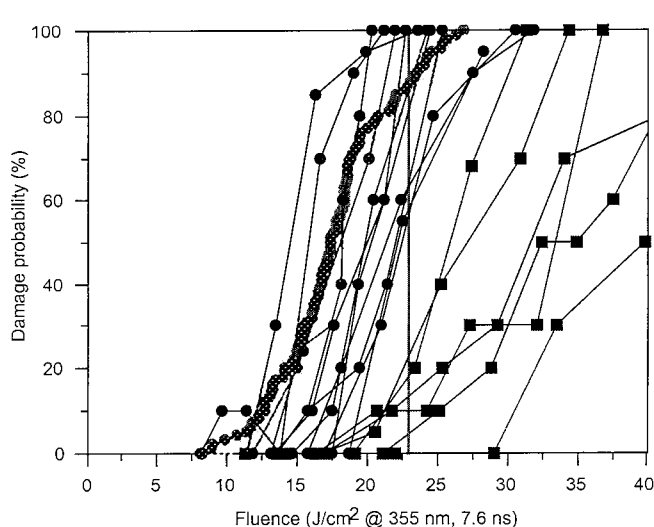


Figure 5. Summary of S/1 (unconditioned) results for all z-cut damage samples from Table 1. Low-T boules (squares) are systematically higher than high-T boules (circles).

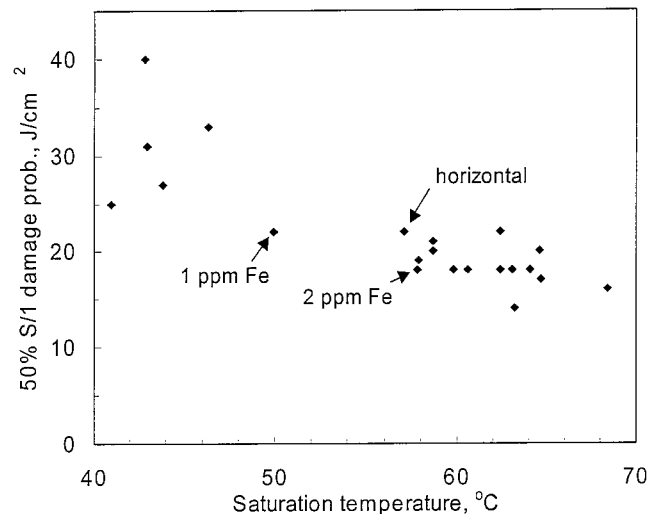


Figure 6. Relationship between saturation temperature and the S:1 50% damage probability fluence for z-cut damage samples

Finally, some production salts are manufactured with EDTA (ethylene diamine tetraacetic acid) as a complexing agent to keep trivalent impurities, particularly transition metals, in solution when the salt is precipitated. Depending on the synthesis details, varying amounts of EDTA are captured in the salt. Boule #'s 731, 737, and 746 grown from EDTA-containing salt in a polycarbonate tank are within the range of others grown at high temperature in glass tanks without EDTA (e.g., #'s 651, 710, 778). On the other hand, boule #'s 756 and 757 grown in glass tanks with EDTA salt are the lowest two of those grown at high temperature, so there could be a slight effect. Boule #727 grown at low temperature in a polycarbonate tank with EDTA salt is within the range of other low-temperature boules grown in glass tanks without EDTA (e.g., #'s 636, 694, 709). Also, addition of 2 ppm of iron (KDP solid basis) in a growth solution containing 0.0057 mol% EDTA did not degrade the damage performance compared to other high-temperature boules, which suggests that EDTA extends the range of acceptable iron contamination in the growth solution.

Based on the results of these experiments, a 50-cm boule (FD-10) was grown to test whether lower growth temperatures might enable NIF-size boules to meet NIF 3 $\omega$  damage requirements. A 1000-L crystallizer was linked by two continuous filtration units to a second Pyrex 1000-L tank, and the boule was grown as the solution temperature was lowered from 42 to 34°C. Six tripler-cut damage samples were taken from the boule representing early to late prism and pyramid material. As shown in Figure 7, the damage results are close to the specification. All but one R/1 profiles overlap DKDP-11 for the first 10-20% damage probability, which is most critical. However, they increase more rapidly to 100% damage probability and, therefore, would be more sensitive to beam fluence spatial modulation. The damage resistance of this large boule did not increase as much as expected based on the small boule experiments, possibly because of the much lower effective filtration rate.

Material balance calculations and chemical analyses were used to estimate the segregation coefficient for iron incorporation into the prismatic sector of the crystals and how it varies with temperature, as shown in Table 2. In both cases, the average of the initial and final solution analyses were used. For the material balance calculations, negligible pyramidal uptake and equal masses of prismatic and pyramidal material were assumed. For the chemical measurement method, the average of at least two prismatic locations were used. Both methods involve assumptions and therefore give only qualitative results. However, the assumptions are different in the two methods, and the close correspondence between them indicates that the basic trend—i.e., that the segregation coefficient differs by about a factor of two between the two temperature ranges—is valid. The temperature trend is also clearly evident directly in the chemical analyses. The iron content in the growth solution is typically about the same before and after the growth run at high temperature but drops by 1/3 in the low temperature runs. The trend is the same in polycarbonate and glass tanks, indicating that tank leaching is a minor issue. Similar trends were observed for Al, but they are not as reliable because of the greater contribution of tank dissolution.

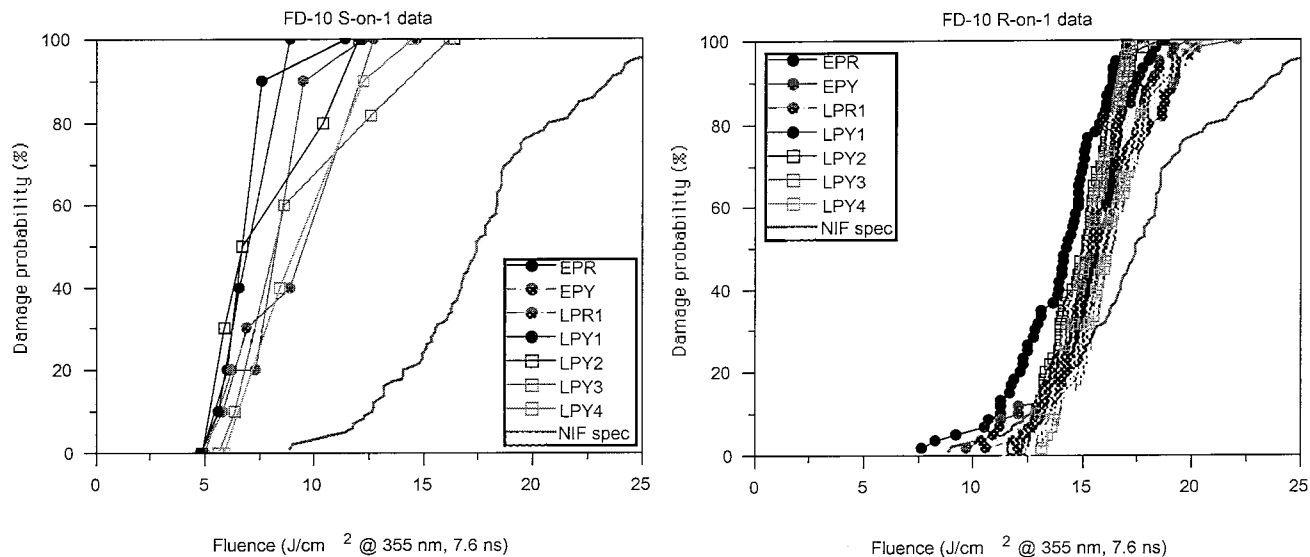


Figure 7. S/1 and R/1 bulk damage profiles for tripler damage samples from low-temperature LLNL boule FD-10. The laser-conditioned (R/1) damage probability is close to but not clearly acceptable for full fluence operation on the NIF.

Table 2. Prismatic segregation coefficients for iron calculated from mass balance and direct measurement of crystals.

	Material balance method			Chemical analysis method		
High T, no EDTA	solution	crystal	coeff. <sup>a</sup>	solution	crystal	coeff. <sup>b</sup>
avg 651, 710, 715, 778	128	133	2.1			
651				96	145	1.5
778				200	740	3.7
Low T, no EDTA						
avg 636, 694, 709, 727, 701, 720	119	291	4.9			
636				123	765	6.2
694				76	507	6.7

<sup>a</sup>assumes that all iron is absorbed into the prism and the prism represents 50% of the crystal.

<sup>b</sup>assumes that the average measured solution and prism Fe/KDP ratios are representative of all growth time

In the presence of EDTA, the material balance calculations indicate that little Fe is incorporated into the crystal except for the high Fe cases of crystals 767 and 768. The latter growth runs had 0.0057 and 0.0019 mole % EDTA/KDP, respectively. Even though this is about a tenfold excess of EDTA (0.001 mole % EDTA corresponds to 4100 ppb Fe in KDP), the EDTA complexation is apparently not strong enough to prevent Fe incorporation into the crystal. A more complete picture is given by the chemical analysis, which detected small amounts of iron in some of the other crystals. Figure 8 shows the segregation coefficient as a function of the EDTA/Fe molar ratio in the solution. The incorporation of Fe into the crystal appears to drop roughly with the logarithm of the EDTA/Fe ratio. A similar result was found for Al in the high temperature regime, and those results are shown in Figure 9.



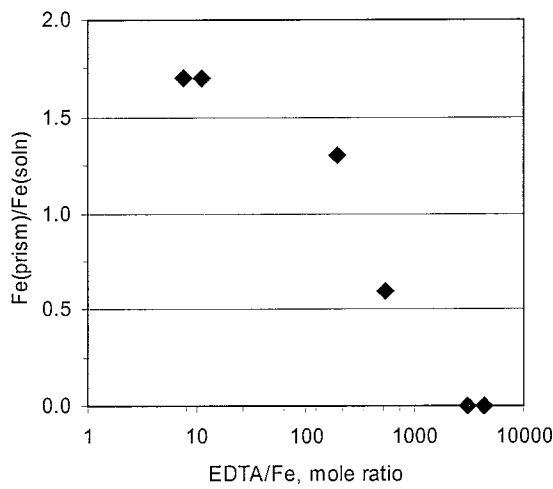


Figure 8. Incorporation of iron into the prismatic sector decreases with the logarithm of the EDTA/Fe molar ratio.

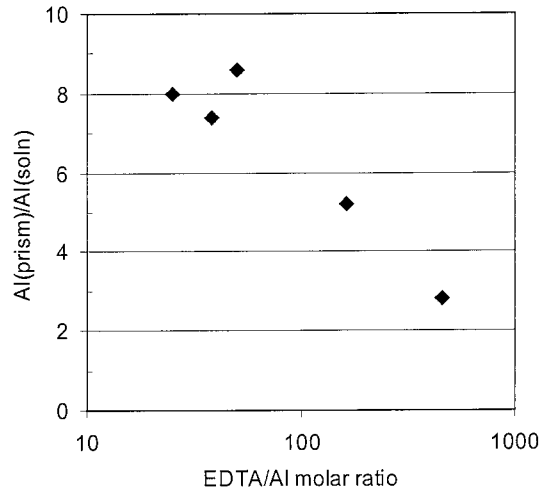


Figure 9. Incorporation of aluminum into the prismatic sector decreases with the logarithm of the EDTA/Al ratio.

Iron incorporation into the crystal increases the optical absorbance in the UV. Yan et al. (unpublished results, 1997) showed that iron is the only important UV-absorbing cation in typical growth solutions and that the absorbance is linear with iron concentration in the crystal. The optical absorbance of some of our crystals was measured at 350 nm using an Ar-ion based photometer, and the relationship between transmittance loss and iron concentration in the crystal is shown in Figure 10. Surface fogging prevented an accurate determination of absorption in many of the crystals, and a linear fit constrained to zero absorbance with no iron found no significant difference between the ordinary (z and tripler cuts) and extraordinary (tripler cuts) optical axes. The iron concentration corresponding to 1% absorbance, the maximum allowed for DKDP used on the National Ignition Facility, occurs at about 250 ppb Fe. Using the segregation coefficients from Table 1, the maximum Fe concentration allowed into the growth solution would be about 100 ppb at high temperature and about 50 ppb at the low temperature. EDTA can easily increase the allowable iron contamination in the growth solution by an order of magnitude.

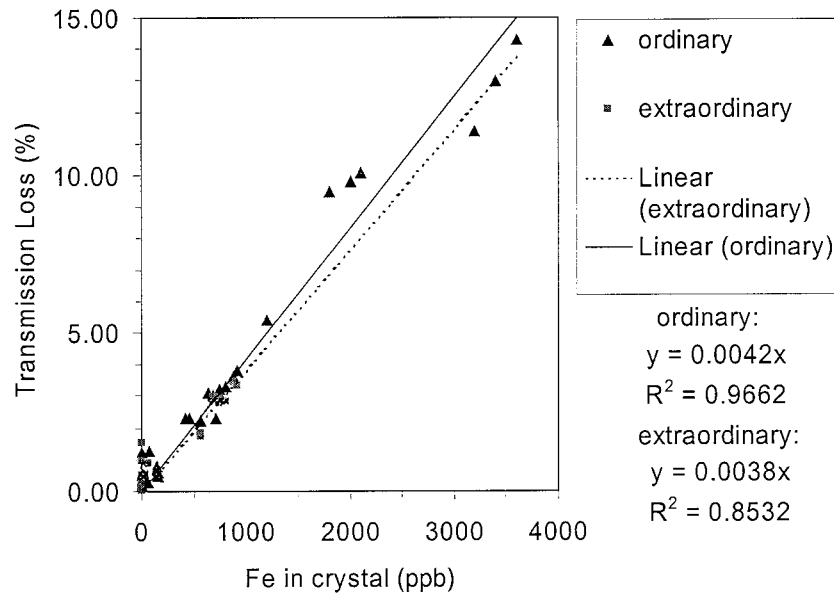


Figure 10. Relationship between transmittance loss at 350 nm and iron concentration in z-cut and type I doubler-cut rapid-growth DKDP crystals.

## 4. DISCUSSION

We currently have no good explanation for the effect of temperature on the damage resistance of the DKDP crystals, although we do have several hypotheses. Concerning the proposal that absorbing nanoparticles are the offending initiation site, there are three possibilities for how temperature might affect their abundance. Since DKDP solubility increases with temperature and since the growth runs are conducted at similar supersaturations, if the impurities leading to nanoparticles have a lower dependence of solubility on temperature, they could be closer to their saturation temperature, hence more prone to precipitation of nanoparticles. Alternatively, or in addition, the higher temperature might accelerate the kinetics of formation of nanoparticles and result in a higher steady-state concentration caused by the competition between formation and filtration. If the sticking coefficient for particulates on the crystal surface becomes smaller at lower temperatures, perhaps fewer particles might be incorporated into the crystal. The higher growth temperature might result in more intrinsic defects in the crystal, perhaps related to the formation of microscopic monoclinic phases. These defects could either cause an absorbing defect in their own right or lead to areas of local stress. The local stress could either directly enhance the damage susceptibility of the crystal or make possible some local harmonic generation that enhances absorption. Finally, there may be a temperature-dependent shedding of particles from the filter. Of course, we have no evidence to substantiate any of these hypotheses, so they need to be systematically investigated and evaluated.

To be useful for the NIF, the beneficial effect of low temperature growth must be demonstrated in a crystallizer capable of growing boules large enough to yield 42-cm plates. Since the mechanism is not clearly understood, and since potentially important parameters (such as the time for container dissolution and the rate of filtration relative to tank volume) are different in 20-L and NIF-size crystallizers, successful implementation of low temperature growth as a method to achieve acceptable damage resistance for rapid growth material is not assured. However, several important steps testing that possibility are in progress. First, the use of a supplemental tank to provide enough dissolved salt at a 45°C saturation temperature to grow a NIF-size boule in a 1000-L crystallizer has been demonstrated. Second, a prototype 1400-L polycarbonate tank to eliminate tank dissolution has been fabricated and has grown a NIF-size KDP crystal. Third, it is conceivable that the number of filtration units associated with the large tanks could be increased to make the filtration rate more similar to that in a 20-L tank. The resolution of these issues will take another year of effort.

While low temperature growth appears to have a beneficial effect on 3 $\omega$  laser-induced bulk damage, there are possible negative consequences that need to be considered. Iron is incorporated into the prism to a greater extent at the lower growth temperatures, leading to higher UV absorbance. For example, there is ~3% absorbance loss at 350-nm in the prism sector of sample #636, which exceed the NIF specification of 1% maximum. In the previous section, we calculated a two- to threefold increase in the Fe segregation coefficient as the growth temperature decreases from the 65-45°C range to the 54-25°C range—about 6 in the low temperature range and 2-3 in the high temperature range. The high temperature value corresponds to the value of 2.3 reported by Zaitseva et al.<sup>5</sup> (note: the prism and pyramid column labels are switched in Table 1 of reference 5). A recent paper suggested that the segregation coefficient may depend on temperature in this way.<sup>10</sup> If verified, this means that meeting the 350-nm absorbance and homogeneity specifications will be more difficult for low temperature growth, although either residual EDTA or the use of large polycarbonate tanks currently being fabricated probably will solve the problem. More data will be required to resolve this issue.

In addition, initial observations suggest that surface crazing is more prevalent in the pyramid sectors of low-temperature rapid-growth DKDP (M. Runkel and L. Chase, unpublished results, 2000). The surface crazing appears to be caused by surface stress induced when water from the atmosphere exchanges with heavy water in the first 50-100  $\mu$ m of the surface over a period of weeks to months. We currently have no good explanation as to why this effect would be greater for material grown at lower temperatures. One hypothesis is that crazing is more prevalent in higher purity crystals with more perfect lattice structures. The craze fractures tend to stop at the pyramid-prism boundary, and the prism is known to have higher levels of impurities. Thermodynamic arguments suggest that stoichiometric preferences are enhanced as temperature decreases. Iron is preferentially incorporated into the prism sector during growth, but it is preferentially excluded from the pyramid sector. The previous paragraph suggested that Fe incorporation into the prism increased as temperature decreases. If preferences truly are enhanced at lower temperatures as suggested recently<sup>10</sup>, impurity concentrations would be lower in the pyramid, possibly explaining the increased tendency for surface crazing. More work is needed on this topic, also.

## CONCLUSIONS

A parametric study of readily controllable process variables suggests that growth temperature is the most important one for current production salt purity. Rapid-growth crystals grown between 45 and 25°C are 1.5 to 2 times more damage resistant than crystals grown between 65 and 45°C. The reason for this improvement is not understood. Extrapolation to NIF-scale boules has been partially successful, and improvements in filtration rate and tank composition are likely to achieve the desired improvement. However, the low-temperature growth appears to have some detrimental effects, such as increasing the tendency for surface crazing upon exchange of surface deuterium with hydrogen in ambient humidity as well as increasing the incorporation of Fe into the prism sector, thereby increasing UV absorbance and optical inhomogeneity between the prism and pyramid sectors. Additional experiments are required to resolve these issues.

## ACKNOWLEDGMENTS

This work was supported under the auspices of the U.S. Department of Energy by the University of California Lawrence Livermore National Laboratory under contract No. W-7405-ENG-48. It received technical support by many staff from the NIF crystal growth and optics metrology groups, particularly Natalia Zaitseva for her contributions to the crystal growth and Tim Sarginson for the optical absorbance measurements.

## REFERENCES

- <sup>1</sup> L. J. Atherton, F. Rainer, J. J. De Yoreo, I. M. Thomas, N. Zaitseva, F. De Marco, "Thermal and laser conditioning of production- and rapid-growth KDP and KD\*P crystals," *Laser-Induced Damage in Optical Materials: 1993*, SPIE Vol. 2114, pp. 36-44 (1993).
- <sup>2</sup> N. P. Zaitseva, L. N. Rashkovich, S. V. Bogatyreva, "Stability of  $\text{KH}_2\text{PO}_4$  and  $\text{K}(\text{H,D})_2\text{PO}_4$  solutions at fast crystal growth rates," *J. Crystal Growth* 148, 276-282 (1995).
- <sup>3</sup> K. E. Montgomery and F. P. Milanovich, "High laser damage threshold KDP crystals," *J. Appl. Phys.* 68, 3979-3982 (1990).
- <sup>4</sup> M. Runkel, M. Yan, J. De Yoreo, and N. Zaitseva, "The effect of impurities and stress on the damage distributions of rapidly grown KDP crystals," *Laser-induced Damage in Optical Materials: 1997*, SPIE Vol. 3244, pp. 211-222 (1997).
- <sup>5</sup> N. Zaitseva, L. Carman, I. Smolsky, R. Torres, and M. Yan, "The effect of impurities and supersaturation on the rapid growth of KDP crystals," *J. Crystal Growth* 204, 512-524 (1999).
- <sup>6</sup> M. Runkel and A. Burnham, "Tripler vs. z-cut bulk damage in KDP and DKDP at  $3\omega$ ," *Proceedings of this symposium* (2001).
- <sup>7</sup> M. Yan, R. Torres, M. Runkel, B. Woods, I. Hutcheon, N. Zaitseva, and J. De Yoreo, "Impurity and laser-induced damage in the growth sectors of rapidly grown KDP crystals," *Laser-induced Damage in Optical Materials: 1996*, SPIE Vol. 2966, pp. 11-16 (1996).
- <sup>8</sup> R. Sharp III and M. Runkel, "Automated damage onset analysis techniques applied to KDP damage and the Zeus small area damage test facility," *Laser-induced Damage in Optical Materials: 1999*, SPIE Vol. 3902, pp. 361-368 (2000).
- <sup>9</sup> M. Staggs, M. Runkel, and M. Yan, "Laser raster conditioning of KDP crystals using XeCl and Nd:YAG Lasers," *Proceedings of this symposium* (2001).
- <sup>10</sup> K. Sangwal and T. Palczynska, "On the supersaturation and impurity concentration dependence of segregation coefficient in crystals grown from solutions," *J. Crystal Growth* 212, 522-531 (2000).